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(56) Documents cited

US 4518575 A US 3816609 A

(58) Field of search

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(54) Method of producing carbon fibers having coil-like filaments

(57) A carbon fiber, which comprises coil-like carbon filaments sometimes together with linear carbon filaments, is efficiently produced by vapor phase pyrolysis of either carbon monoxide gas or an aliphatic hydrocarbon gas, preferably acetylene, at 300–1000°C in the presence of a transition metal such as, e.g., Fe, Co, Ni or Cr and a compound of P, As, Sb, Bi, S, Se or Te, preferably a phosphorus compound, e.g. PCl_3 , or a sulfur compound, e.g. H_2S . The carbon-containing gas may be mixed with hydrogen gas and/or a diluent gas. Each of the obtained coil-like filaments is 0.05–5 μm in diameter and is coiled such that the coil outer diameter is 2 to 10 times the filament diameter and such that the number of turns per length of 10 μm is from 5/D to 50/D, where D is the coil outer diameter in μm .



FIG.1

10 μm

GB 2 248 230 A

FIG.1

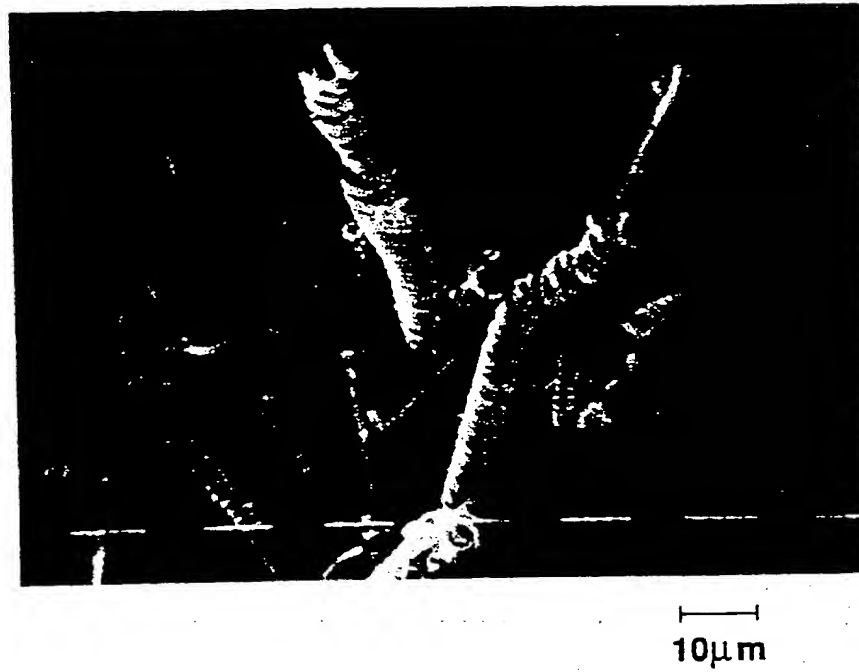
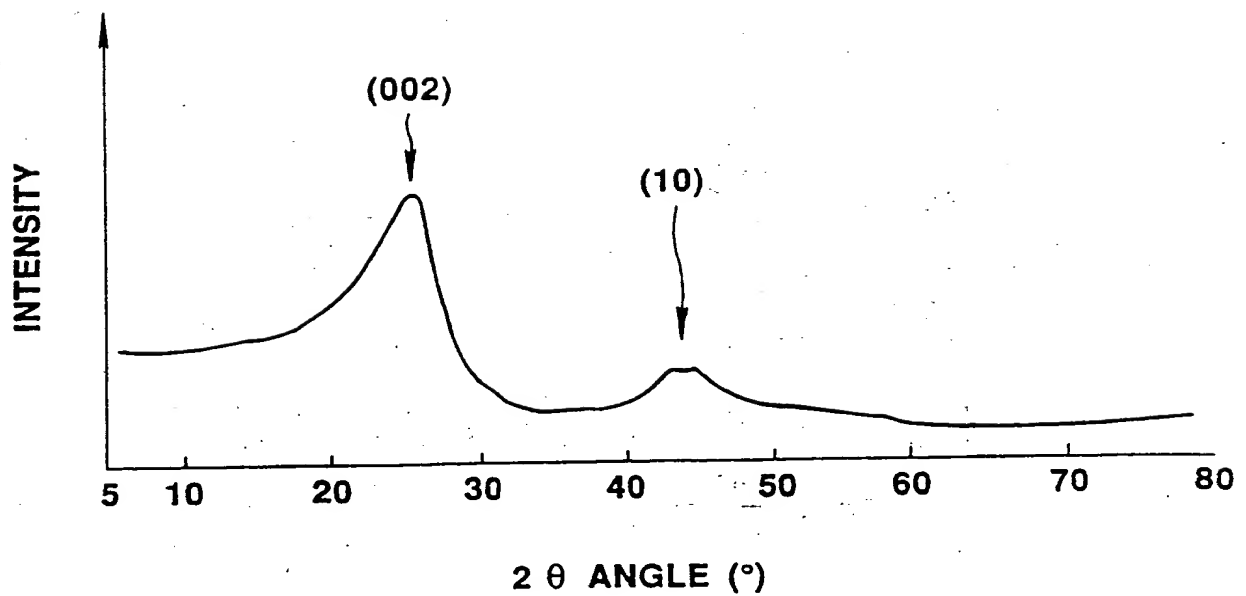


FIG.2



METHOD OF PRODUCING CARBON FIBERS HAVING
COIL-LIKE FILAMENTS

This invention relates to a method of producing a carbon fiber having coil-like filaments.

5 Carbon fibers are very high in specific strength and hence have been used as basic or reinforcing components of composite materials. Besides, it is under study to utilize other properties of carbon fibers such as electrical conductivity, heat conductivity and
10 adsorptive ability in some functional materials.

Carbon fibers are commonly produced by charring or graphitizing an organic fibrous raw material called precursor. Typical precursor materials are polyacrylonitrile and pitch. Also it is known to form carbon
15 fibers by vapor phase pyrolysis of a hydrocarbon. For example, JP 51-33210 shows producing carbon fibers by passing a mixture of an aliphatic or aromatic hydrocarbon gas and a carrier gas through a tube kept heated at 1030-1300°C first at a relatively high flow rate to
20 create nuclei for growth of carbon filaments and thereafter at a lower flow rate suitable for growth of filaments. Although there are many variations in the particulars of known methods for producing carbon fibers, the obtained carbon fibers always consist of
25 linear filaments.

In GB 2,233,971 A, we have disclosed a unique carbon fiber comprising coil-like filaments each of

which is an essentially carbon filament 0.05 to 5 μm in diameter and is coiled such that the coil outer diameter is 2 to 10 times as large the filament diameter and such that the number of turns per length of 10 μm is in the
5 range from $5/D$ to $50/D$, where D represents the coil diameter in μm . The coil-like filaments are, for example, about 100 to 1000 μm in length. According to the earlier application a carbon fiber having such coil-like filaments is produced by subjecting an aliphatic
10 hydrocarbon gas, preferably acetylene gas, to vapor phase pyrolysis at a temperature in the range from 300 to 1000°C in the presence of a transition metal such as Fe, Co, Ni or Cr. In most cases the obtained coil-like carbon fiber has a considerable amount of linear
15 filaments together with coil-like filaments.

Our subsequent studies have revealed that by the method disclosed in GB 2,233,971 A it is not always successful to efficiently produce carbon filaments coiled in the above described manner.

20 SUMMARY OF THE INVENTION

It is an object of the invention to provide a method for efficiently producing a carbon fiber having coil-like filaments as disclosed in the earlier application.

25 According to the invention there is provided a method of producing a carbon fiber having coil-like filaments, the method comprising subjecting a carbon-

containing gas selected from carbon monoxide gas and aliphatic hydrocarbon gases to vapor phase pyrolysis at a temperature in the range from 300 to 1000°C in the presence of a transition metal and a compound of a Group V or VI element selected from phosphorus, arsenic, antimony, bismuth, sulfur, selenium and tellurium.

The gist of the present invention is to carry out the vapor phase pyrolysis of either carbon monoxide gas or an aliphatic hydrocarbon gas in the presence of a compound of a specifically selected element of Group V or VI together with a transition metal. By using such a compound the efficiency of forming beautifully coiled carbon filaments is greatly improved.

By the method according to the invention it is possible to efficiently produce a carbon fiber comprising coil-like filaments each of which is an essentially carbon filament 0.05 to 5 μm in diameter and is beautifully coiled such that the coil outer diameter is 2 to 10 times as large as the filament diameter and such that the number of turns per length of 10 μm is in the range from 5/D to 50/D, where D represents the coil outer diameter in μm .

In this method it is preferred to use acetylene as the carbon-containing gas, Ni, Co, Fe or Cr as the transition metal and phosphorus or sulfur as the Group V or VI element. The carbon-containing gas may be mixed with hydrogen gas and/or an inactive diluent gas.

A carbon fiber produced by a method according to the invention may comprise linear filaments together with the coil-like filaments.

Carbon fibers produced by a method according to the invention are useful for various purposes almost similarly to conventional short fibers of carbon. For example, a composite material having high strength, wear resistance and low friction coefficient is obtained by filling interstices between the filaments of a carbon fiber obtained by the invention with a carbon matrix formed by a known method. The coil-like filaments afford a resilient or cushioning property to the composite material and/or contribute to enhanced toughness of the composite material by firmly gripping the carbon matrix. Besides, the carbon fibers obtained by the invention are useful as adsorbents, filters, electrode materials for batteries, etc.

Furthermore, the spring properties of the beautifully coiled carbon filaments can be utilized in cushioning materials and micromechanical elements which are required to exhibit spring properties even at high temperatures and/or in corrosive atmospheres. Also it is possible to utilize the coil-like carbon filaments, which are electrically conductive, in switching elements to control the flow of a current by stretching or contracting to produce a change in contact area.

In the accompanying drawings:

Fig. 1 is a SEM photographs of an example of coil-like carbon fibers produced by a method according to the invention; and

5 Fig. 2 is a chart showing the X-ray diffraction pattern of an example of coil-like carbon fibers produced by a method according to the invention.

In the present invention either carbon monoxide or an aliphatic hydrocarbon is used as the carbon source
10 for producing carbon fibers. In the case of a hydrocarbon, either a saturated hydrocarbon such as methane, ethane, propane or butane or an unsaturated hydrocarbon such as acetylene, ethylene or propylene can be used. Benzene is not usable because vapor phase pyrolysis of
15 benzene, which occurs at temperatures above 1000°C, does not give a coil-like carbon fiber. In this invention acetylene is particularly preferred mainly because transition metals exhibit very good catalytic effects on the fiber forming pyrolysis of acetylene.

20 It is optional and rather preferable to mix the selected carbon-containing gas with hydrogen gas. By varying the amount of addition of hydrogen gas it is possible to control the temperature for the vapor phase pyrolysis of and consequently control the coil-like
25 shape of the deposited carbon filaments. It is unfavorable to use more than 10 mols of hydrogen per mol of carbon-containing compound because of excessively

suppressing the pyrolysis of the carbon-containing compound. It is preferable that the molar ratio of hydrogen to the carbon-containing compound falls in the range from 1:1 to 5:1.

5 Also it is optional to use an inactive diluent gas such as, for example, argon, helium or nitrogen irrespective of the addition of hydrogen gas. This is another means to control the coil-like shape of the deposited carbon filaments. It is suitable that the
10 molar ratio of the diluent gas to the carbon-containing gas is not more than 10:1, and a preferred range of the molar ratio is from 1:1 to 5:1.

 The pyrolytic reaction of the carbon-containing gas must be carried out at a temperature in the range from
15 300 to 1000°C. At temperatures below 300°C it is difficult to fully decompose the carbon-containing compound. When the reaction temperature is higher than 1000°C, only linear filaments of carbon are formed. A preferred range of the reaction temperature is from 400
20 to 900°C.

 It is suitable to carry out the pyrolytic reaction under the atmospheric pressure or under a slightly reduced pressure not lower than 200 mmHg. At a pressure above the atmospheric pressure or lower than 200 mmHg it
25 is difficult to suitably control the pyrolytic reaction.

 The vapor phase pyrolysis of a selected hydrocarbon needs to be carried out in the presence of a transition

metal which serves as a catalyst. Without using any transition metal it is very difficult to form carbon fibers. For example, the transition metal can be selected from Fe, Co, Ni, Cr, W, Ti, Nb and Mo and
5 alloys of these metals. In general it is preferable to use Ni, Co, Fe or Cr.

It is possible to use a sheet or plate of a selected transition metal as a catalytic substrate on which carbon fibers deposit. In such a case it is desirable
10 to polish the transition metal substrate surface for the deposition. There are some other ways to introduce a transition metal catalyst into the reaction system. For example, a powder of the transition metal or a salt (e.g., nitrate or chloride) of the metal is scattered on
15 a substrate of a different material or in a suitable section of the reactor, or a solution of such a salt of the transition metal is applied to a substrate of a different material, or an organic compound of the transition metal (e.g., ferrocene) is introduced into
20 the reactor together with the hydrocarbon gas. In the case of using a compound of a transition metal it is necessary to choose a compound which readily decomposes at the temperature employed for the pyrolysis reaction of the carbon-containing gas because it is intended to
25 carry out the pyrolysis reaction in the presence of a transition metal in metallic form. Therefore, in the case of using a transition metal compound it is

preferable to feed hydrogen gas together with the carbon-containing gas so that the transition metal compound may easily be reduced to the metal.

When it is desired to accomplish the above-described pyrolysis at a relatively low temperature within the aforementioned range, it is necessary to use a powder of a transition metal as catalyst. In powder form the transition metal catalyst exhibits very high activity so that the decomposition of the carbon-containing gas to deposit carbon in the form of coil-like filaments will be eased even at a relatively low temperature.

It is necessary that together with a transition metal either a compound of a Group V element selected from P, As, Sb and Bi or a compound of a Group VI element selected from S, Se and Te should be present in the reaction system for thermally decomposing the carbon-containing gas. Examples of useful compounds are PH_3 , PCl_3 , PF_3 , PF_5 , RPH_2 (R represents a lower alkyl group; usually methyl or ethyl), R_2PH , P_4S_3 , PR_3 , AsH_3 , AsCl_3 , AsF_3 , AsF_5 , AsR_3 , SbH_3 , SbCl_3 , SbF_3 , SbF_5 , SbR_3 , BiH_3 , BiCl_3 , BiF_3 , BiF_5 , BiR_3 , H_2S , RSH , R_2S , R_2S_2 , S_2Cl_2 , SCl_2 , H_2Se , RSeH , R_2Se , Se_2Cl_2 , SeCl_2 , H_2Te , RTeH , R_2Te , Te_2Cl_2 and TeCl_2 . It is preferable to use a phosphorus compound or a sulfur compound to obtain coil-like carbon fiber filaments at good yield.

There are two ways of introducing a selected compound of a Group V or VI element into the reaction system. The compound will be referred to as the catalytic compound.

5 The first way is treating a selected transition metal with the catalytic compound at an elevated temperature in advance of the pyrolytic reaction of a carbon-containing gas. In practice it is suitable to accomplish the treatment by first placing the transition
10 metal in the reactor for the pyrolytic reaction and then introducing a gas of the catalytic compound into the reactor while maintaining the reactor nearly at the temperature at which the pyrolytic reaction is to be carried out. In most cases it suffices to make this
15 treatment for a short period of time ranging from several minutes to tens of minutes.

 The second way is introducing a gas of the catalytic compound into the reactor together with a carbon-containing gas and, optionally, hydrogen gas
20 and/or an inactive gas. In this case it suffices that the concentration of the catalytic compound in the mixed gas is from 1 to 1000 ppm. For efficiently producing coil-like carbon fiber filaments it is unsuitable to unnecessarily increase the concentration of the
25 catalytic compound.

 In the presence of a transition metal and a compound of a Group V or VI element, the object of the

pyrolytic reaction of a carbon-containing gas can be accomplished by continuing the reaction for tens of minutes to several hours.

The following nonlimitative examples are
5 illustrative of the invention.

EXAMPLE 1

A quartz tube 40 mm in inner diameter and 1000 mm in length was used as a reaction tube for a thermal CVD process, and 2.5 g of nickel powder was put into a
10 lengthwise central section of the reaction tube so as to spread over a length of 150 mm. The central section of the reaction tube was inserted in an electric furnace, and the temperature in the furnace was gradually raised up to 700°C while argon gas was passed through the tube.

15 The temperature in the furnace was kept at 700°C, and, in place of argon gas, hydrogen sulfide (H_2S) gas was passed through the tube for 5 min at a constant rate of 1.7 ml/min. Then the feed of hydrogen sulfide gas was stopped, and acetylene gas (50 ml/min) and argon gas
20 (50 ml/min) were passed through the reaction tube under the atmospheric pressure. This operation was continued for 1 hr. As a result, 2.0 g of carbon in the form of very short fiber filaments deposited on the nickel powder in the central and forward sections of the tube
25 where the temperature was 500-700°C during the pyrolysis of acetylene gas. The deposited carbon fiber was a mixture of about 50 wt% of coil-like filaments and about

50 wt% of linear filaments. Fig. 1 is a SEM photograph of coil-like carbon filaments produced in this example.

The coil-like carbon filaments obtained in Example 1 were 0.1 to 2 μm in filament diameter, about 200 μm in length, 100 to 2000 in aspect ratio and 0.2 to 20 μm in outer diameter of coil. With respect to each coil-like filament, the ratio of coil outer diameter to filament diameter was in the range from 2:1 to 10:1, and the number of turns of the coil per length of 10 μm was 5 to 20 times the reciprocal of the coil outer diameter (μm).

EXAMPLE 2

Using the same apparatus as in Example 1, 2.5 g of nickel powder was spread in the central section of the reaction tube. The central section of the reaction tube was inserted in an electric furnace in which the temperature was maintained at 750°C. Using argon gas as a carrier gas, hydrogen gas and acetylene gas containing hydrogen sulfide gas were passed through the reaction tube under the atmospheric pressure. The flow rates of the respective gases were as follows.

Acetylene:	30 ml/min
Hydrogen sulfide:	0.05 ml/min
Hydrogen:	70 ml/min
Argon:	40 ml/min

Therefore, in the mixed gas the concentration of H_2S was about 350 ppm.

The above operation was continued for 2 hr. As a result, 3.2 g of carbon in the form of very short fibers deposited on the nickel powder in the central and forward sections of the reaction tube where the temperature was 520-750°C during the pyrolysis operation. The deposit contained about 40 wt% of coil-like carbon filaments. The obtained coil-like carbon filaments were 0.1 to 1 μm in filament diameter, about 300 μm in length, 300 to 3000 in aspect ratio and 0.5 to 20 μm in coil outer diameter. With respect to each coil-like filament, the ratio of coil outer diameter to filament diameter was in the range from 5:1 to 20:1, and the number of turns of the coil per length of 10 μm was 5 to 30 times the reciprocal of the coil outer diameter (μm).

The coil-like carbon filaments were ground in an agate mortar, and the resultant powder was subjected to X-ray diffraction analysis with Cu-K α line at 40 kV, 30 mA. Fig. 2 shows the obtained X-ray diffraction chart. The peak of the (002) diffracted lines is at 2θ angle of 24.9°, and the broadness (7°) of the half-width is indicative of low crystallinity of the carbon filaments.

EXAMPLE 3

Using the same apparatus as in Example 1, a nickel substrate 20 mm in width, 1000 mm in length and 3 mm in thickness was placed in the reaction tube. The central section of the reaction tube was inserted in an electric

furnace in which the temperature was maintained at 700°C. In a buffer tank methyl mercaptan gas was diluted with argon gas to 1/10000 by volume, and acetylene gas and the argon gas containing methyl mercaptan were passed through the reaction tube at the following flow rates, respectively.

Acetylene: 50 ml/min

Argon: 15 ml/min

Therefore, the concentration of methyl mercaptan in the mixed gas was about 80 ppm.

The above operation was continued for 30 min. As a result, 3.0 g of carbon in the form of very short fibers deposited on the nickel substrate in the central and forward sections of the reaction tube, where the temperature was 650-700°C during the pyrolysis operation. Most of the deposited carbon filaments were coil-like filaments. The coil-like carbon filaments were 0.5 to 1 μm in filament diameter, about 1000 μm in length, 1000 to 2000 in aspect ratio and 1 to 20 μm in coil outer diameter. With respect to each coil-like filament, the ratio of coil outer diameter to filament diameter was in the range from 2:1 to 10:1, and the number of turns of the coil per length of 10 μm was 5 to 30 times the reciprocal of the coil outer diameter (μm). By X-ray diffraction analysis the coil-like filaments proved to be carbon filaments low in crystallinity.

EXAMPLE 4

The process of Example 2 was modified by using phosphorus trichloride PCl_3 in place of hydrogen sulfide, increasing the quantity of nickel powder to 3.0 g and lowering the temperature in the electric furnace to 700°C . Acetylene gas containing PCl_3 , hydrogen gas and argon gas were passed through the reaction tube at the following flow rates, respectively.

	Acetylene:	30 ml/min
10	Phosphorus trichloride:	0.05 ml/min
	Hydrogen:	70 ml/min
	Argon:	40 ml/min

Therefore, the concentration of PCl_3 in the mixed gas was about 350 ppm.

15 The above operation was continued for 2 hr. As a result, 4.0 g of carbon in the form of very short fibers deposited on the nickel powder in the central and forward sections of the reaction tube. The deposit contained about 80 wt% of coil-like filaments. The
20 coil-like carbon filaments were 0.2 to 1 μm in filament diameter, about 200 μm in length, 200 to 1000 in aspect ratio and 0.4 to 10 μm in coil outer diameter. With respect to each coil-like filament the ratio of coil outer diameter to filament diameter was in the range
25 from 2:1 to 10:1, and the number of turns of the coil per length of 10 μm was 3 to 30 times the reciprocal of the coil outer diameter (μm).

EXAMPLE 5

The process of Example 4 was modified only in that the quantity of nickel powder was increased to 3.5 g, the temperature in the electric furnace was raised to 800°C and that the feed rate of PCl_3 was decreased to 0.01 ml/min without varying the flow rates of acetylene gas, hydrogen gas and argon gas. In this case the concentration of PCl_3 in the mixed gas was about 70 ppm.

The pyrolysis operation was continued for 2 hr. As a result 4.0 g of carbon in the form of very short fibers deposited on the nickel powder in the central and forward sections of the reaction tube. The deposit contained about 40 wt% of coil-like filaments. The coil-like carbon filaments were 0.1 to 1 μm in filament diameter, about 300 μm in length, 300 to 3000 in aspect ratio and 0.2 to 10 μm in coil outer diameter. With respect to each coil-like filament the ratio of coil outer diameter to filament diameter was in the range from 2:1 to 10:1, and the number of turns of the coil per length of 10 μm was 5 to 50 times the reciprocal of the coil outer diameter (μm).

EXAMPLE 6

The process of Example 5 was modified only in that phosphorus trifluoride PF_3 was used in place of PCl_3 . Acetylene gas containing PF_3 , hydrogen gas and argon gas were passed through the reaction tube at the following flow rates, respectively.

Acetylene: 30 ml/min
Phosphorus trifluoride: 0.09 ml/min
Hydrogen: 70 ml/min
Argon: 40 ml/min

5 Therefore, the concentration of PF_3 in the mixed gas was about 640 ppm.

The pyrolysis operation was continued for 2 hr. As a result, 3.8 g of carbon in the form of very short fibers deposited on the nickel powder in the central and forward sections of the reaction tube. The deposit
10 contained about 20 wt% of coil-like filaments. The coil-like carbon filaments were 0.1 to 1 μm in filament diameter, about 200 μm in length, 200 to 2000 in aspect ratio and 0.2 to 10 μm in coil outer diameter. With
15 respect to each coil-like filament the ratio of coil outer diameter to filament diameter was in the range from 2:1 to 10:1, and the number of turns of the coil per length of 10 μm was 5 to 50 times the reciprocal of the coil outer diameter (μm).

20

25

CLAIMS

1 A method of producing a carbon fiber comprising coil-like filaments, the method comprising subjecting a carbon-containing gas selected from carbon monoxide gas and aliphatic hydrocarbon gases to vapor phase pyrolysis at a temperature in the range from 300 to 1000°C in the presence of a transition metal, a salt thereof or an alloy thereof, and a compound of an element selected from phosphorus, arsenic, antimony, bismuth, sulfur, selenium and tellurium.

10 2. A method according to Claim 1, wherein said temperature is in the range from 400 to 900°C.

3. A method according to Claim 1 or 2, wherein said carbon-containing gas is acetylene gas.

15 4. A method according to Claim 1, 2 or 3, wherein said transition metal is selected from Fe, Co, Ni, Cr, W, Ti, Nb and Mo and their alloys.

5. A method according to any one of the preceding claims wherein said transition metal is in the form of a solid.

20 6. A method according to any one of claims 1 to 4, wherein said transition metal is in the form of a powder.

7. A method according to any of Claims 1 to 6, wherein said compound is a phosphorus compound selected from PH_3 , PCl_3 , PF_3 , PF_5 , RPH_2 , R_2PH , PR_3 and P_4S_3 , wherein each R individually represents a lower alkyl group.

25

8. A method according to Claim 7, wherein each R

represents methyl or ethyl.

9. A method according to any of Claims 1 to 6,
wherein said compound is a sulfur compound selected from
 H_2S , S_2Cl_2 , SCl_2 , RSH , R_2S , R_2S_2 and P_4S_3 , wherein each R
5 individually represents a lower alkyl group.

10. A method according to Claim 9, wherein each R
represents methyl or ethyl.

11. A method according to any of Claims 1 to 10,
wherein said transition metal is treated with said compound
10 prior to the vapor phase pyrolysis of said carbon-
containing gas.

12. A method according to any of Claims 1 to 10,
wherein said compound is mixed with said carbon-containing
gas prior to pyrolysis.

15 13. A method according to any of the preceding
claims, wherein the vapor phase pyrolysis of said carbon-
containing gas is carried out in the presence of hydrogen
gas.

14. A method according to Claim 13, wherein the
20 molar ratio of said hydrogen gas to said carbon-containing
gas is in the range from 1:1 to 5:1.

15. A method according to any of the preceding
claims, wherein the vapor phase pyrolysis of said carbon-
containing gas is carried out in the presence of an
25 inactive diluent gas.

16. A method according to Claim 15, wherein the

molar ratio of said inactive diluent gas to said carbon-containing gas is in the range from 1:1 to 5:1.

17. A method according to any of the preceding claims, wherein said vapor phase pyrolysis is carried out
5 under the atmospheric pressure.

18. A method according to any of Claims 1 to 16, wherein said vapor phase pyrolysis is carried out under a reduced pressure not lower than 200 mmHg.

19. A method of producing a carbon fiber
10 comprising coil-like filaments, substantially as hereinbefore described in any of Examples 1 to 6.

Patents Act 1977
Examiner's report to the Comptroller under
Section 17 (The Search Report)

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Relevant Technical fields

(i) UK CI (Edition K) C1A (AJ4)

(ii) Int CI (Edition 5) D01F

Databases (see over)

(i) UK Patent Office

(ii)

Search Examiner

C A CLARKE

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29 JULY 1991

Documents considered relevant following a search in respect of claims

1 to 19

Category (see over)	Identity of document and relevant passages	Relevant to claim(s)
X	US 4518575 (PHILLIPS PETROLEUM) see whole document	1 to 7 and 11 to 17 at least
Y	US 3816609 (ESSO) see Claim 1	1 at least

SF2(p)

GPZAAQ

Category	Identity of document and relevant passages	Relevant to claim(s)

Categories of documents

X: Document indicating lack of novelty or of inventive step.

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